Phase separation of electrons strongly coupled with phonons in cuprates and manganites.

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Abstract

Recent advanced Monte Carlo simulations have not found superconductivity and phase separation in the Hubbard model with on-site repulsive electron-electron correlations. We argue that microscopic phase separations in cuprate superconductors and colossal magnetoresistance (CMR) manganites originate from a strong electron-phonon interaction (EPI) combined with unavoidable disorder. Attractive electron correlations, caused by an almost unretarded EPI, are sufficient to overcome the direct inter-site Coulomb repulsion in these charge-transfer Mott-Hubbard insulators, so that low energy physics is that of small polarons and small bipolarons (real-space electron (hole) pairs dressed by phonons). They form clusters localised by disorder below the mobility edge, but propagate as the Bloch states above the mobility edge. I identify the Fröhlich finite-range EPI with optical phonons as the most essential for pairing and phase separation in superconducting layered cuprates. The pairing of oxygen holes into heavy bipolarons in the paramagnetic phase (current-carrier density collapse (CCDC)) explains also CMR of doped manganites due to magnetic break-up of bipolarons in the ferromagnetic phase. Here I briefly present an explanation of high and low-resistance phase coexistence near the ferromagnetic transition as a mixture of polaronic ferromagnetic and bipolaronic paramagnetic domains due to unavoidable disorder in doped manganites.

Keywords: electron-phonon interaction, phase separation, bipolarons, cuprates, manganites

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I. INTRODUCTION

There are still many theories that attempt to explain the phenomenon of high-temperature superconductuctivity in cuprates and other related materials. In general, the pairing mechanism of carriers could be not only "phononic" as in the BCS theory [1](left-hand upper corner in Fig.(1)) or its strong-coupling bipolaronic extension [2, 3] (right-hand upper corner in Fig.(1)) , but also "excitonic" [4, 5], "plasmonic" [6, 7], "magnetic" [8, 9], "kinetic" [10], or purely "coolombic" due to a mirror-nested Fermi surface [11]. The BCS theory like any mean-field theory is rather universal, so that it describes well the cooperative quantum phenomenon of superconductivity even with these non-phononic mechanisms, if the coupling is weak (left-hand lower corner in Fig.(1)). The main motivation behind these concepts is that high superconducting critical temperature, T_c, could be achieved by replacing phonons in the conventional BCS theory by higher frequency bosonic modes, such as plasmons, spin waves (pseudomagnons), or even by the direct Coulomb repulsion combined with unconventional pairing symmetries.

Actually, following original proposal by P. W. Anderson [12] , many authors [13, 14] assumed that the electron-electron interaction in novel superconductors is very strong but repulsive and it provides high T_c without any phonons (right-hand lower corner in Fig.(1)). A motivation for this concept can be found in the earlier work by Kohn and Luttinger[15], who showed that the Cooper pairing of repulsive fermions is possible. However the same work clearly showed that T_c of repulsive fermions is extremely low, well below the mK scale. Nevertheless, the BCS and BCS-like theories (including the Kohn-Luttinger consideration) heavily rely on the Fermi-liquid model of the *normal* state, which fails in many high-temperature superconductors. If the normal state is not the Fermi-liquid, then there is no direct reason to reject the assumption. In fact there is little doubt that strong onsite repulsive correlations (Hubbard U) are an essential feature of the cuprates. Indeed all undoped cuprate compounds are *insulators* with the insulating gap about $2eV$ or so. But if the repulsive correlations are weak, one would expect a metallic behaviour of a half-filled d-band of copper in cuprates, or, at most, a much smaller gap caused by lattice and spin distortions (i.e. due to charge and/or spin density waves [16, 17]). It is a strong onsite repulsion of d-electrons in cuprates which results in their parent insulating "Mott" state. When onsite correlations are strong and dimensionality is low, there is an alternative to the

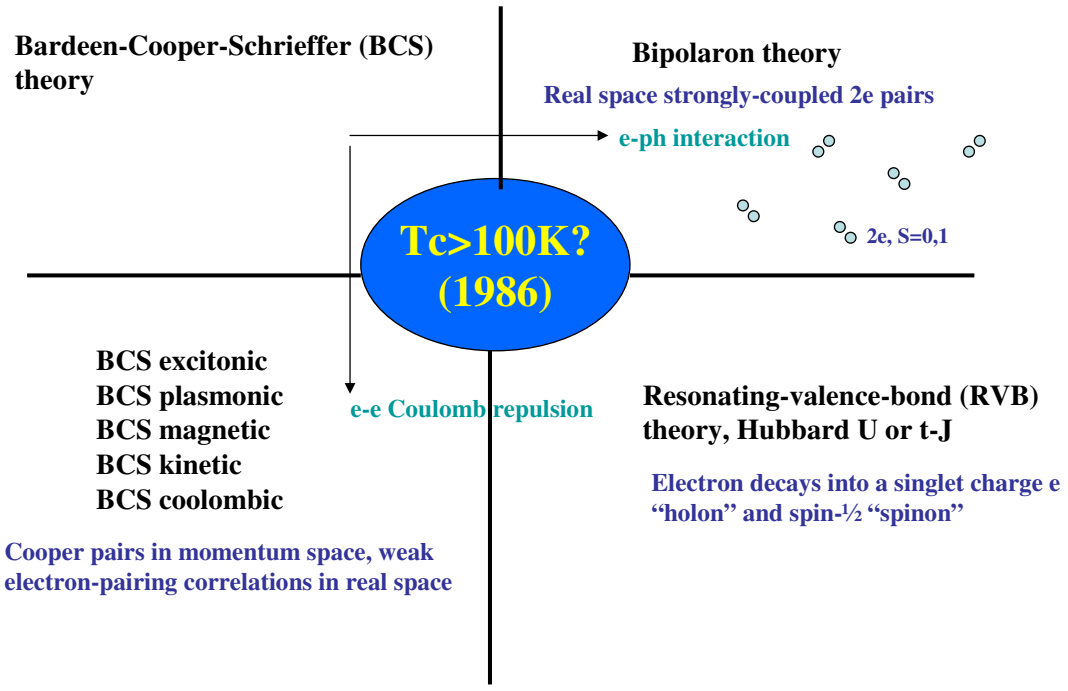

FIG. 1: A few theories of high-temperature superconductivity. The highest T_c is predicted in the BCS to bipolaron crossover [3] for the phonon pairing mechanism (upper half of the diagram). Lower half of the diagram represents a number of non-phononic mechanisms of pairing.

usual Fermi-liquid. In Anderson's resonating-valence-bond (RVB) model [12] the ground state supports "topological solitons" (the so-called spinons and holons), such as occur in one-dimensional Hubbard model. Theoretically holons could be paired by a superexchange interaction without any additional glue like phonons or spin-waves [18].

To discriminate one theory with respect to another one has to rely on experimental facts and/or on exact theoretical results. Some variational Monte Carlo (VMC) simulations with a (projected) BCS-type trial wave function (see, for example [14] and references therein), and a number of other analytical and numerical studies appeared to back up superexchange pair-

ing. However recent studies by Aimi and Imada [19], using an advanced sign-problem-free Gaussian-Basis Monte Carlo (GBMC) algorithm have shown that these variational methods, as well as other approximations, overestimated the normal state energy and therefore overestimated the condensation energy by at least an order of magnitude, so that the Hubbard model does not account for high-temperature superconductivity. The ground state of the model is a normal Fermi liquid with no superconductivity and no stripes. This remarkable result is in line with earlier numerical studies using the auxiliary-field quantum (AFQMC) [20] and constrained-path (CPMC) [21] Monte-Carlo methods, none of which found superconductivity in the Hubbard model.

On the other hand compelling experimental evidence for a strong EPI has arrived from isotope effects [22], high resolution angle resolved photoemission spectroscopies (ARPES) [23], a number of optical [24], neutron-scattering [25, 26], tunnelling [27] spectroscopies of cuprates, and from recent pump-probe experiments [28]. I have suggested that a strong long-range Fröhlich EPI is the key to both high-temperature superconductivity and colossal magnetoresistance [29]. Our [30, 31, 32, 33] and some other studies [34, 35, 36] of strongly-coupled polarons and bipolarons have shown that the long-range discrete Fröhlich EPI in the presence of the strong Coulomb repulsion does not lead to an enormous enhancement of the carrier effective mass characteristic of the Hubbard-Holstein model (HHM), and could provide a higher (room temperature) superconductivity [37].

II. PHASE SEPARATION IN CUPRATES

In the strong-coupling regime, where the BCS electron-phonon coupling constant is relatively large, $\lambda \gtrsim 1$, pairing is individual [2], in contrast with the collective Cooper pairing [1]. Bipolarons survive even in the normal state above their Bose-Einstein condensation temperature representing a simplest "cluster" of carriers. While the Fröhlich and Coulomb interactions alone could not lead to larger clusters like strings or stripes [40], shorter-range interactions as the deformation [41], Holstein [42], Jahn-Teller [43, 44] or a strong nonlinear [45] EPIs could favor bound states of more than two carriers.

Formation of polaronic clusters can be analytically studied in the strong-coupling regime in the framework of a generic "Fröhlich-Coulomb" model (FCM) [2, 30, 32]. The model Hamiltonian explicitly includes a long-range electron-phonon and the Coulomb interactions

as well as the kinetic and deformation energies. The implicitly present large Hubbard U term prohibits double occupancy and removes the need to distinguish fermionic spins since the exchange interaction is negligible compared with the direct Coulomb and the electron-phonon interactions in complex oxides.

Introducing fermionic, $c_{\mathbf{n}}$, and phononic, $d_{\mathbf{m}\alpha}$, operators the FCM Hamiltonian is written as

$$\begin{aligned} H = & -\sum_{\mathbf{n}\neq\mathbf{n}'}\left[T(\mathbf{n}-\mathbf{n}')c^{\dagger}_{\mathbf{n}}c_{\mathbf{n}'}-\frac{1}{2}V_c(\mathbf{n}-\mathbf{n}')c^{\dagger}_{\mathbf{n}}c_{\mathbf{n}}c^{\dagger}_{\mathbf{n}'}c_{\mathbf{n}'}\right] \\ & -\sum_{\alpha,\mathbf{nm}}\omega_\alpha g_\alpha(\mathbf{m}-\mathbf{n})(\mathbf{e}_\alpha\cdot\mathbf{u}_{\mathbf{m}-\mathbf{n}})c^{\dagger}_{\mathbf{n}}c_{\mathbf{n}}(d^{\dagger}_{\mathbf{m}\alpha}+d_{\mathbf{m}\alpha}) \\ & +\sum_{\mathbf{m}\alpha}\omega_\alpha\left(d^{\dagger}_{\mathbf{m}\alpha}d_{\mathbf{m}\alpha}+1/2\right), \end{aligned} \tag{1}$$

where $T(\mathbf{n})$ is the hopping integral in a rigid lattice, $\mathbf{e}_\alpha$ is the polarization vector of the αth vibration coordinate, $\mathbf{u}_{\mathbf{m}-\mathbf{n}}\equiv(\mathbf{m}-\mathbf{n})/|\mathbf{m}-\mathbf{n}|$ is the unit vector in the direction from electron $\mathbf{n}$ to ion $\mathbf{m}$, $g_\alpha(\mathbf{m}-\mathbf{n})$ is the dimensionless EPI function, and $V_c(\mathbf{n}-\mathbf{n}')$ is the inter-site Coulomb repulsion. $g_\alpha(\mathbf{m}-\mathbf{n})$ is proportional to the *force* $f_{\mathbf{m}}(\mathbf{n})$ acting between the electron on site $\mathbf{n}$ and the ion on $\mathbf{m}$. For simplicity we assume that all phonon modes are non-dispersive with frequencies ω_α and include spin in the definition of $\mathbf{n}$ (here $\hbar=1$).

Bipolarons on a two-dimensional lattice of ideal octahedra (that can be regarded as a simplified model of the copper-oxygen perovskite layer) have been studied in Ref. [32]. Due to poor screening, the hole-ion interaction was taken as purely coulombic,

$$g_\alpha(\mathbf{m}-\mathbf{n})=\frac{\kappa_\alpha}{|\mathbf{m}-\mathbf{n}|^2},$$

where $\alpha=x,y,z$ with $\kappa_x=\kappa_y=\kappa_z/\sqrt{2}$ accounting for the experimental fact that c-axis (z-polarized) phonons couple to in-plane holes stronger than others. The direct hole-hole repulsion is

$$V_c(\mathbf{n}-\mathbf{n}')=\frac{V_c}{\sqrt{2}|\mathbf{n}-\mathbf{n}'|},$$

so that the repulsion between two holes in the nearest neighbour (NN) configuration is V_c. The nearest neighbour hopping T_{NN}, the next-nearest neighbour (NNN) hopping across copper T_{NNN} and the hopping between the pyramids T'_{NNN} have been included, Fig.(2).

The polaron level shift in this model is given by the lattice sum,

$$E_p=2\kappa_x^2\omega_0\sum_{\mathbf{m}}\left(\frac{1}{|\mathbf{m}-\mathbf{n}|^4}+\frac{h^2}{|\mathbf{m}-\mathbf{n}|^6}\right)=31.15\kappa_x^2\omega_0, \tag{2}$$

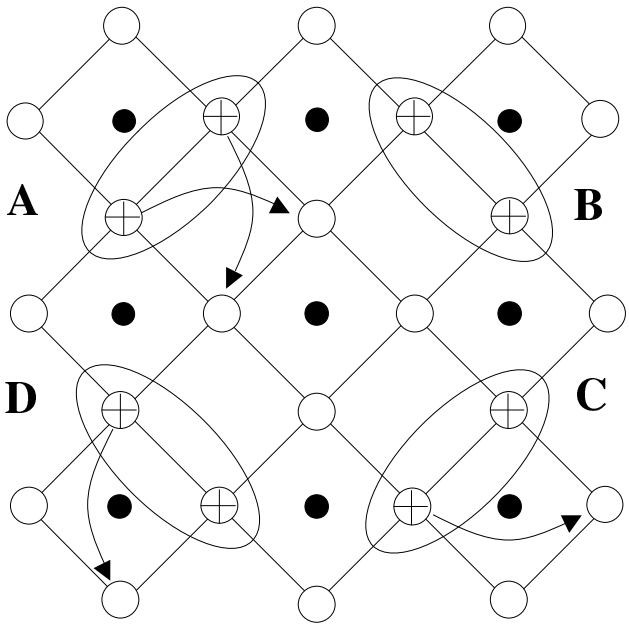

FIG. 2: Four degenerate in-plane bipolaron configurations A, B, C, and D . Some single-polaron hoppings are indicated by arrows. (Reproduced from [32], (c) IoP, 2002.)

where the factor 2 accounts for two layers of apical sites, and the in-plane lattice constant is $a = 1$ and $\omega_\alpha = \omega_0$. For reference, the Cartesian coordinates are $\mathbf{n} = (n_x + 1/2, n_y + 1/2, 0)$, $\mathbf{m} = (m_x, m_y, h)$, and n_x, n_y, m_x, m_y are integers. The polaron-polaron attraction is

$$V_{ph}(\mathbf{n} - \mathbf{n}') = 4\omega_0 \kappa_x^2 \sum_{\mathbf{m}} \frac{h^2 + (\mathbf{m} - \mathbf{n}') \cdot (\mathbf{m} - \mathbf{n})}{|\mathbf{m} - \mathbf{n}'|^3 |\mathbf{m} - \mathbf{n}|^3}. \tag{3}$$

Performing the lattice summations for the NN, NNN, and NNN$'$ configurations one finds $V_{ph} = 1.23\, E_p$, $0.80\, E_p$, and $0.82\, E_p$, respectively. As a result, we obtain a net inter-polaron interaction as $v_{NN} = V_c - 1.23\, E_p$, $v_{NNN} = \frac{V_c}{\sqrt{2}} - 0.80\, E_p$, $v'_{NNN} = \frac{V_c}{\sqrt{2}} - 0.82\, E_p$, and the mass renormalization exponents (see below) as $g^2_{NN} = 0.38(E_p/\omega)$, $g^2_{NNN} = 0.60(E_p/\omega)$ and $(g'_{NNN})^2 = 0.59(E_p/\omega)$. At $V_c > 1.23\, E_p$, no bipolarons are formed and the system is a polaronic Fermi liquid. Polarons tunnel in the *square* lattice with the renormalised hopping integrals $t = T_{NN} \exp(-0.38 E_p/\omega)$ and $t' = T_{NNN} \exp(-0.60 E_p/\omega)$ for NN and NNN hoppings, respectively. The polaron mass is $m^* \propto 1/(t + 2t')$.

If $V_c < 1.23\, E_p$, then intersite NN bipolarons form. The intersite bipolarons tunnel in the plane via four resonating (degenerate) configurations A, B, C, and D, as shown in Fig.(2). In the first order of the renormalised hopping integral, one should retain only these lowest energy configurations and discard all the processes that involve configurations with higher

energies. These inter-site bipolarons already move in the *first* order of the single polaron hopping. This remarkable property is entirely due to the strong on-site repulsion and long-range electron-phonon interactions that leads to a non-trivial connectivity of the lattice. This fact combines with a weak renormalization of t' yielding a *superlight* bipolaron with the mass $m^{**} \propto \exp(0.60\, E_p/\omega)$. We recall that in the Holstein model $m^{**} \propto \exp(2E_p/\omega)$ [2]. Thus the mass of the small Fröhlich bipolaron in the perovskite layer scales approximately as a *cubic root* of that of the Holstein bipolaron.

At even stronger EPI, $V_c < 1.16E_p$, NNN bipolarons become stable. More importantly, holes can now form 3- and 4-particle clusters. The dominance of the potential energy over kinetic in the transformed Hamiltonian enables us to readily investigate these many-polaron cases. Three holes placed within one oxygen square have four degenerate states with the energy $2(V_c - 1.23E_p) + V_c/\sqrt{2} - 0.80E_p$. The first-order polaron hopping processes mix the states resulting in a ground state linear combination with the energy $E_3 = 2.71V_c - 3.26E_p - \sqrt{4t^2 + t'^2}$. It is essential that between the squares such triads could move only in higher orders of polaron hopping. In the first order, they are immobile. A cluster of four holes has only one state within a square of oxygen atoms. Its energy is $E_4 = 4(V_c - 1.23E_p) + 2(V_c/\sqrt{2} - 0.80E_p) = 5.41V_c - 6.52E_p$. This cluster, as well as all bigger ones, are also immobile in the first order of polaron hopping. Hence a strong EPI combined with the Coulomb repulsion could cause clustering of polarons into finite-size mesoscopic textures. Importantly QMC studies of mesoscopic textures [44] including lattice deformations and the Coulomb repulsion show that pairs (i.e. bipolarons) dominate over phase separation since they effectively repel each other [2]. I would like to stress that at distances much larger than the lattice constant the polaron-polaron interaction is always repulsive, and the formation of infinite clusters, stripes or strings is prohibited [40].

III. PHASE SEPARATION IN FERROMAGNETIC MANGANITES

The conventional double-exchange (DEX) model of the ferromagnetism and colossal magnetoresistance (CMR), proposed half a century ago and generalized more recently to include the electron-phonon interaction, is in conflict with a number of contemporary experiments [38, 39]. Among those experiments are site-selective spectroscopes, which show that oxygen p-holes are current carriers, rather than d-electrons in ferromagnetic manganites [46].

Also, some ferromagnetic manganites manifest an insulating-like optical conductivity at all temperatures, contradicting the DEX notion that their ferromagnetic phase is metallic [47]. CMR is observed in manganese pyrochlores [48] where DEX is non-existent.

On the other hand, the pairing of oxygen holes into heavy bipolarons in the paramagnetic phase (current carrier-density collapse (CCDC)) and their magnetic break-up in the ferromagnetic phase is compatible with the above and many other observations explaining CMR, isotope effects, and pseudogaps observed in doped manganites [22, 49, 50, 51, 52]. Different from other models CCDC predicted the first-order phase transition, now firmly established in single CMR crystals [53]. CCDC was directly observed in the Hall effect at the ferromagnetic transition [54].

More recently we have proposed an explanation of high and low-resistive phase mixing near the ferromagnetic transition observed in tunnelling [55] and some other experiments as the mixture of polaronic ferromagnetic domains and bipolaronic paramagnetic domains due to unavoidable disorder in doped manganites [39]. Using the fact that the phase transition in a homogeneous system is of the first order in a wide range of parameters [38, 49] one can average the magnetisation, $\sigma(T)$, with the Gaussian distribution of random transition temperatures T_{Ci}s caused by disorder around the experimental Curie temperature T_C to obtain

$$\sigma(T) = \frac{1}{2}\mathrm{erfc}\left(\frac{T - T_C}{\Gamma}\right), \tag{4}$$

where Γ is the distribution width and $\mathrm{erfc}(z) = (2/\pi^{1/2})\int_z^\infty dy \exp(-y^2)$. The CCDC with disorder, Eq. (4) fits nicely the experimental magnetizations near the transition with physically reasonable Γ of the order of 10K, depending on doping, Fig.(3). Hence, we believe that the random distribution of transition temperatures with the width Γ across the sample caused by the randomness of the bipolaron binding energy is responsible for the phase coexistence near the transition as seen in the tunnelling experiments [55].

Resistivity of inhomogeneous two-phase systems has to be calculated numerically. Nevertheless, the comprehensive numerical simulations are consistent with a simple analytical expression for the resistivity of the binary mixture,

$$\rho = \rho_1^{1-\nu}\rho_2^{\nu}, \tag{5}$$

which is valid in a wide range of the ratios ρ_1/ρ_2 [56]. Here $\rho_{1,2}$ is the resistivity of each phase, respectively, and ν is the volume fraction of the second phase.

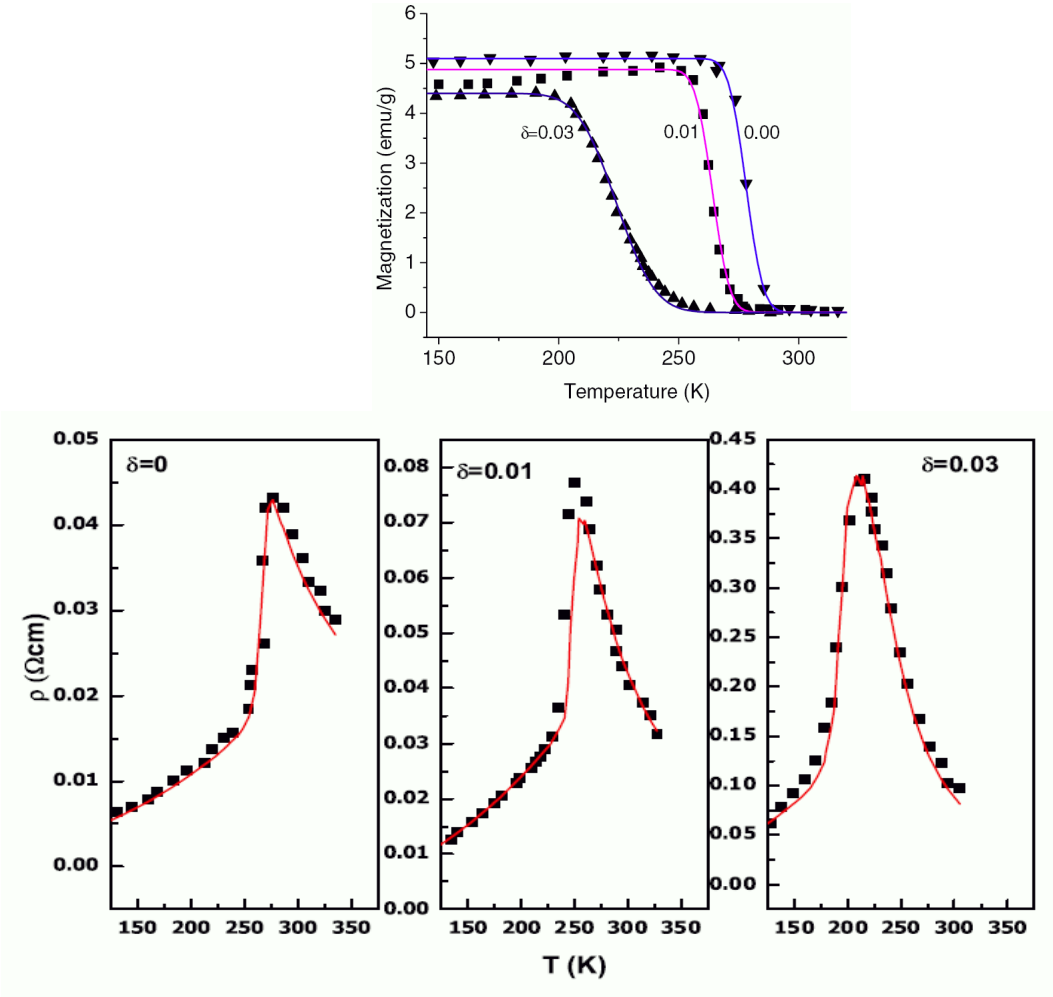

FIG. 3: CCDC model (lines) describes the experimental magnetisation (upper panel) and resistivity (lower panels) near the ferromagnetic transition in $La_{0.7}Ca_{0.3}Mn_{2-\delta}Ti_{\delta}O_3$ (symbols [57]), if the phase coexistence caused by disorder is taken into account. No fitting parameters are used in Eq.(5) but the experimental resistivity well below and well above the transition and the experimental magnetization. (Reproduced from [39], (c) American Physical Society, 2006.)

In the framework of CCDC, the resistivity of the paramagnetic phase is $\rho_1(T) = f(T)\exp(\Delta/2k_BT)$ and the resistivity of the ferromagnetic phase is $\rho_2(T) = \phi(T)$, where $f(T)$ and $\phi(T)$ are polynomial functions of temperature depending on the scattering mechanisms, and Δ is the bipolaron binding energy. Well below the transition $\phi(T)$ can be parameterized as $\phi(T) = \rho_0 + aT^2$, and $f(T) = bT$ well above the transition, where the tem-

perature independent parameters ρ_0, a, $\Delta/2$ and b are taken directly from the experiment. The volume fraction ν of the ferromagnetic phase is simply the relative magnetization in our model, $\nu = \sigma(T)$, also available from the experiment. As a result, Eq. (5) provides the quantitative description of $\rho(T)$ in the transition region without any fitting parameters by using the experimental resistivity far away from the transition and the experimental magnetization, as shown in Fig.(3). Studies of the low-field magnetoresistance of $Sm_{1-x}Sr_xMnO_3$ (x=0.45) which was sintered at different elevated temperatures followed by fast cooling also found very good qualitative agreement with CCDC [58].

Finally, our concept of polaronic metal in ferromagnetic manganites [38, 51] has been substantiated by the angle-resolved photoemission spectroscopy data for the bilayer manganite $La_1.2Sr_1.8Mn_2O_7$, where a polaron metallic state below T_C has been clearly observed [59].

IV. CONCLUSIONS

For although high-temperature superconductivity has not yet been targeted as '*the shame and despair of theoretical physics*', - a label attributed to conventional superconductivity during the first half-century after its discovery - the parlous state of current theoretical constructions has led to a current consensus that there is no consensus on the theory of high-T_c superconductivity. Nevertheless impressive amount of experimental data (for example [22, 23, 24, 25, 26, 27, 28]) and accurate numerical simulations [19] have ruled out the simple Hubbard model as an explanation of high-temperature superconductivity (lower right-hand corner in Fig.(1)). Our view, which I have briefly presented here in connection with the phase separation, is that the extension of the BCS theory towards the strong interaction between electrons and ion vibrations (upper right corner in Fig.(1)) describes the phenomenon naturally. The high temperature superconductivity exists in the crossover region of the EPI strength from the BCS-like polaronic to bipolaronic superconductivity as was predicted by us [3] before the discovery [60], proposed as an explanation of high T_c in cuprates [61, 62], Fig.(4), and explored in greater detail after the discovery [2, 63, 64, 65, 66, 67].

Bipolarons also explain CMR [38] and, combined with disorder, the phase separation in manganites [39]. The observation of the pseudogap and nodal quasiparticles in colossal magnetoresistive manganites [68] which have been considered as a characteristic feature of the

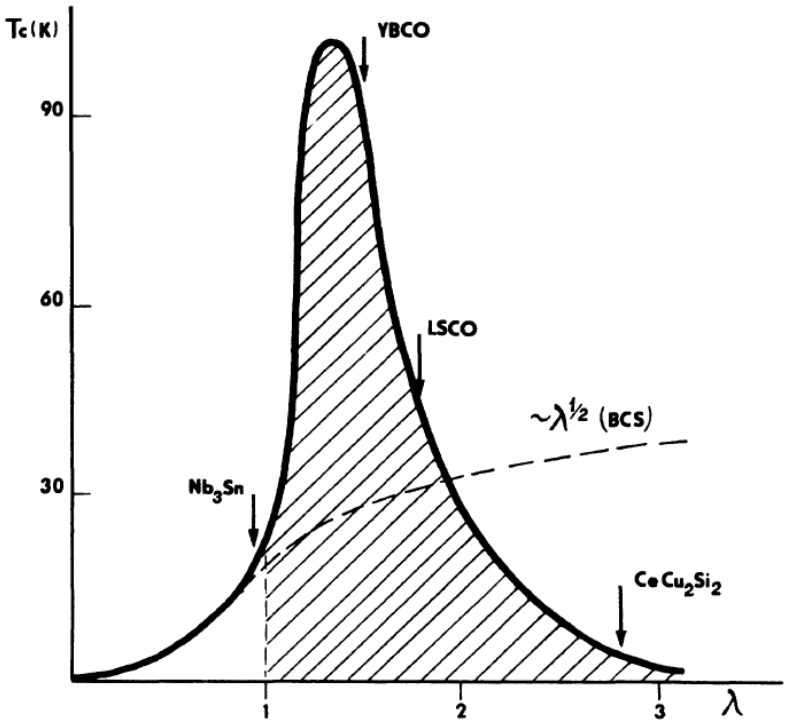

FIG. 4: Dependence of the superconducting critical temperature on EPI coupling constant. The shading shows the (bi)polaronic domain. The dotted line corresponds to the BCS-Eliashberg theory. (Reproduced from [62], (c) American Physical Society, 1988.)

copper oxide, further substantiates our analogy between high-temperature superconducting and CMR oxides.

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